

Measurements of surface ozone at semi-arid site Anantapur (14.62°N, 77.65°E, 331 m asl) in India

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Abstract In the present study, an attempt has been made to examine the governing photochemical processes of surface ozone (O_3) formation in rural site. For this purpose, measurements of surface ozone and selected meteorological parameters have been made at Anantapur (14.62°N, 77.65°E, 331 m asl), a semi-arid zone in India from January 2002 to December 2003. The annual average diurnal variation of O_3 shows maximum concentration 46 ppbv at noon and minimum 25 ppbv in the morning with 1σ standard deviation. The average seasonal variation of ozone mixing ratios are observed to be maximum (about 60 ppbv) during summer and minimum (about 22 ppbv) in the monsoon period. The monthly daytime and nighttime average surface ozone concentration shows a maximum (55 ± 7 ppbv; 37 ± 7.3 ppbv) in March and minimum (28 ± 3.4 ppbv; 22 ± 2.3 ppbv) in August during the study period. The monthly average high (low) O_3 48.9 ± 7.7 ppbv (26.2 ± 3.5 ppbv) observed at noon in March (August) is due to the possible increase in precursor gas concentration by anthropogenic activity and the influence of meteorological parameters. The rate of increase of surface ozone is high (1.52 ppbv/h) in March and lower (0.40 ppbv/h) in July. The average rate of increase of O_3 from midnight to midday is 1 ppbv/h. Surface temperature is highest (43–44°C) during March and April months leading to higher photochemical production. On the other hand, relative humidity, which is higher during the rainy season, shows negative correlation with temperature and ozone mixing ratio. It can be seen that among the two parameters are measured, correlation of surface ozone with wind speed is better ($R^2=0.84$) in compare with relative humidity ($R^2=0.66$).

Keywords Semi-arid zone · Surface ozone · Anthropogenic emissions · Meteorological parameters

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1 Introduction

Atmospheric ozone plays an important role in the physico-chemical processes of the troposphere but it has also strong oxidant properties, which at certain ambient concentration levels may cause damage to humans, animals and vegetation (Finalyson-Pitts and Pitts 1997; Bates, 1994). The problem of tropospheric ozone has changed dimensions about 15 years ago, when it was realized that increased surface ozone is not only a local urban problem, as there is an almost global increase of surface rural ozone concentrations, especially in the northern hemisphere during the twentieth century, which is attributed primarily to photochemical production (Volz and Kley 1988; Kalabokas and Repapis 2004).

Ground-level ozone is formed from its precursors, nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) and non-methane hydrocarbon (NMHC), by complex and non-linear photochemical reactions in sunlight. The potential for ozone formation varies with the ratio of ambient NO_x /NMHC concentrations (Seinfeld 1986). Species that are sensitive to the formation of ozone can be identified by conducting field measurement studies (Sillman et al. 1995), whose results help to determine whether the species that constrains the formation of ozone is NO_x or NMHC. However, various factors such as precursor emissions, atmospheric chemistry, meteorology, and deposition (NRC 1991) may also influence the concentration of ambient ozone. Kondratyev and Varotsos (2001a, b) have pointed out that the prediction of global climate change and of change in the atmosphere's ozone distribution, is impossible without consideration of the complexity of all interactive processes including chemistry and dynamics of the atmosphere. In order to understand the long-range behavior of air pollution, Varotsos (2005a) employed power law relationships (Varotsos and Cartalis 1991).

The existence of a background level of ozone in the atmosphere is well established. There is considerable interest in quantifying surface background ozone concentrations and associated trends, as they serve to define a lower boundary with respect to reductions of ozone by control of anthropogenic precursors. Background ozone is generally defined as the fraction of ozone present in a given area that is not attributed to anthropogenic sources of local origin. As such, background ozone has several well-documented sources, both natural and anthropogenic. These include: (1) downward transport of stratospheric ozone through the free troposphere to near ground level, (2) in situ ozone production from methane emitted from swamps and wetlands reacting with natural oxides of nitrogen (NO_x) (from soils, lightening strikes and downward transport of NO from the stratosphere), (3) in situ production of ozone from reactions of biogenic Volatile Organic Compounds (VOCs) with natural NO_x ($\text{NO} + \text{NO}_2$) and (4) long-range transport of ozone from distant pollutant sources (EPA 1993; Vingarzan and Thompson 2004; Varotsos and Cartalis 1991).

Ozone levels at any location are governed mainly by two factors namely local chemistry and transport (Oltmans and Levy 1994; Jaffe et al. 1998; Lal et al. 1998). Near the earth's surface, ozone is mainly produced by photochemistry involving pollutants which are released from various anthropogenic activities like industrialization, biomass burning, automobile exhausts, etc. (Fishman and Crutzen 1978). Because of the variability in the sources of ozone precursors like CO, CH_4 , NO_x and volatile organic compounds (VOCs) and the interplay of advection and other circulation processes, ozone concentrations exhibit large spatial and temporal variations. Meteorological conditions also affect the transport and mixing of precursor gases. Study of ozone chemistry is important at rural sites because ozone precursors may get transported there from the near-by urban or industrial areas. Many investigators (Varotsos et al. 2001; 2006, Kalabokas et al. 2000; 2004 and Wang et al. 2006) have analyzed the rural ozone levels which were higher than the values observed in Northern Europe and Atlantic Ocean. The purpose of the present paper is to investigate

the seasonal and diurnal cycling of the atmospheric ozone and meteorological parameters at the surface monitored in Anantapur, India during January 2002 to December 2003.

2 Observation site

Surface ozone is measured using an analyzer (O_3 41 M; Environnement s.a., France) has been deployed in the campus of Sri Krishnadevaraya University (SKU), a semi-arid rural site at Anantapur (14.62°N, 77.65°E, 331 m asl). The SKU campus is about 14 km away from the center of the city of Anantapur (Fig. 1). The climate is hot and dry in summer (March–May), hot and humid in monsoon (June–November) and dry in winter (December–February). Normal rainfall is of the order of 450 mm during the whole year. This region receives very little rainfall. The weather at the measurement site during May is very hot due to intense solar radiation. In winter season fair weather condition is prevailing with wind speed of the order of $3\text{--}4\text{ m s}^{-1}$, with southeasterly direction and moderate relative humidity.

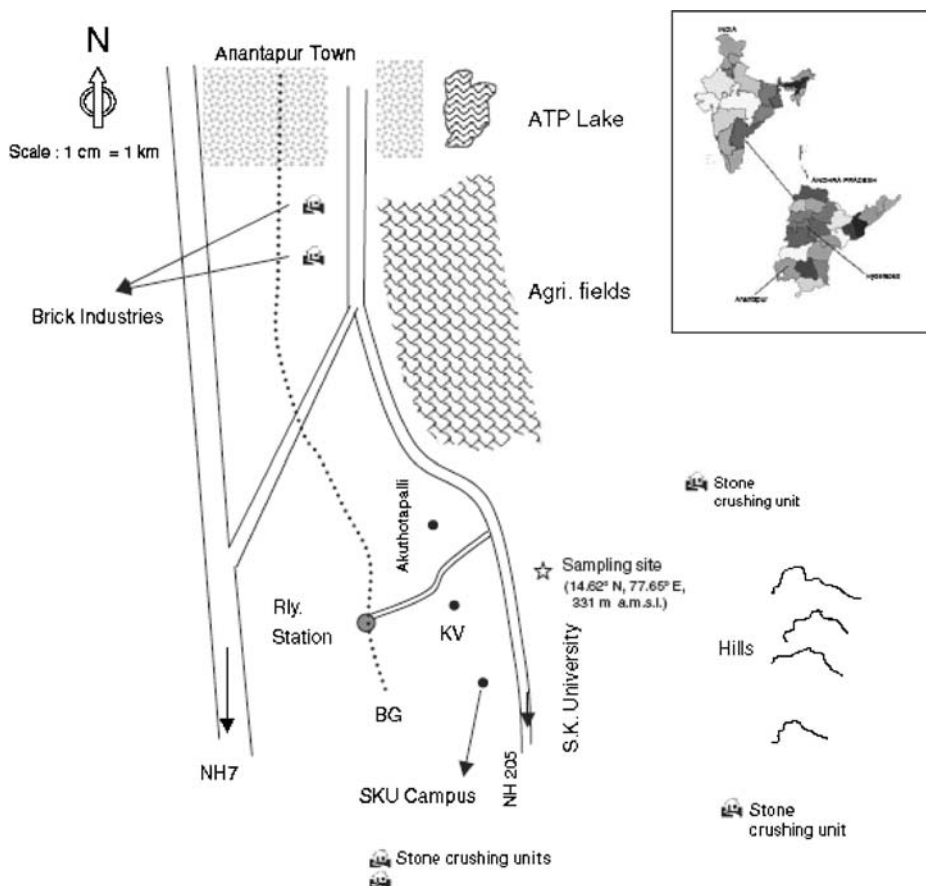


Fig. 1 Observational site (star) and surroundings, showing the position of the University Campus, town area, National Highways, small-scale industries, etc

With in a 50-km radius, the region has a number of cement plants, limekilns, brick making and slab polishing units. These industries release large quantities of particulate matter every day into the atmosphere.

3 Experimental technique

Surface ozone is measured using an analyzer (O_3 41 M; Environnement s.a., France) based on absorption of UV radiation at 253.7 nm by ozone molecules. Contribution by other species in the absorption and scattering of the radiation in the cell is eliminated by comparing the measurement with ozone free air in reference mode. A mercury lamp is the source of this radiation that is absorbed by the ozone molecules present in the ambient air filled in the absorption cell of length of about 60 cm. The absorbed signal as well as the reference signal is measured by a detector. Ozone concentration is estimated using the Beer-Lambert law. This is kept at constant temp and, based on the lamp intensity (by changing the current to the UV lamp), the concentration of O_3 varies. A built-in ozone generator is employed in this instrument. The calibration is carried out until the preset values of reading in the analyzer match the current applied to the UV lamp. The O_3 determination is based on a commercial instrument using UV mercury absorption of 253.7 nm radiation. The O_3 analyzer is so designed that it absorbs ozone at 253.7 nm, and the UV mercury lamp used in the instrument also emits light of the same wavelength. The calibration factor is not be required in this process. The detector is employed before and after the absorption takes place in the fixed length flow path. So the variations in the intensity of the light are balanced. In order to check the zero reading of the analyzer zero air has to be admitted which is free of ozone. If the analyzer is reading a higher value for zero air, then the ozone scrubber in the analyzer needs to be changed; for which the analyzer scrubbers continually check the zero every 10 s and goes to the sample line. These are reformed for 15 min each on a daily basis for an initial period of 1 year and then once very 3–4 weeks afterward. The minimum detection limit of the analyzer is about 1 ppbv and its response is about 10 s. Lal et al. (1998, 2000) and Nair et al. (2002) have also been employed the same O_3 analyzer described above in their study. The absolute accuracy of these types of system is reported to be 5% (Kleinman et al. 1994).

4 General meteorological conditions

A summary of the average prevailing meteorological conditions during the study period is shown in Fig. (4b–f). The monthly mean wind speeds, direction, maximum and minimum of temperature, relative humidity and total rain fall are obtained for the measurement made from Meteorology facility established at the Sri Krishnadevaraya University station. The winds at 1,104 hPa level are basically low in (speed $\sim 3 \text{ ms}^{-1}$) S–SW direction, during the months of January to May. The prevailing winds start shifting from June to August. During this period the wind speeds are high and reaches to a peak value of $5\text{--}6 \text{ m s}^{-1}$ in the months of June to August. The change in the prevailing winds and the mean air temperature increases initially in pre-monsoon months, (March to May) and decreasing in the monsoon periods; and the ambient relative humidity (RH) varies from 75–85% to 40–50%. The southwest monsoon occurs in southern India from June to September when the ITCZ moves northward over India, rearing up to about 30°N during July. The winter, or northeast monsoon, occurs from October to November when the ITCZ moves southward. The winds

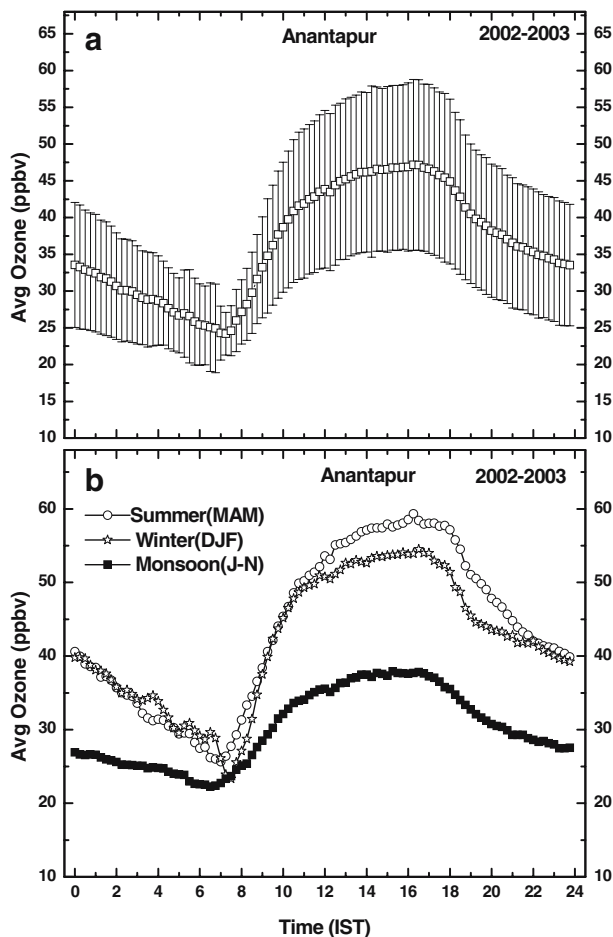
over India are southwesterly to westerly on the southern side of the ITCZ and easterly to north easterly on the northern side of the ITCZ (Asnani 1993).

5 Results and discussions

5.1 Diurnal and seasonal variations of surface ozone

Figure 2a shows the diurnal variation of O_3 averaged over the a period of 2 years from January 2002 to December 2003. The vertical bars denote 1σ standard deviation. The annual average diurnal variations of ozone observed during daytime are high (~ 46 ppbv) and low (~ 25 ppbv) during late night and early morning hours. Mixing ratio of ozone starts increasing at about 07:30 h and it reaches a maximum value at about 1600 hours. The diurnal peak is rather broad, extending from 11:00 to 18:00 hours and the mixing ratio of ozone starts decreasing after 18:00 h. The diurnal cycle of O_3 is explained by nighttime O_3

Fig. 2 Annual average **a** and seasonal average **b** diurnal variations of ozone mixing ratios at Anantapur from January 2002 to December 2003. Vertical bars are in average of 1σ standard deviations



destruction under the formation of a nocturnal inversion layer and by daytime convective boundary layer mixing and photochemical O_3 production (Pochanart et al. 2001). Mixing ratios of ozone decrease more slowly during the evening than they increase during the morning. This has been attributed to slower titration of ozone by NO during late evening time at the rural sites (Naja and Lal 2002; Wang et al. 2006). Generally, urban sites show almost similar morning production rates and evening loss rates (i.e., symmetric variation). Peaks observed in the late afternoon and minima in the early morning hours preceding dawn, which is a typical characteristics resulted from atmospheric photochemical reactions, transport and chemical depletion in rural region (Ahammed et al. 2006; Naja and Lal 2002; Debaje and Kakade 2006). The variation of surface ozone, within a day, may be helpful in delineating the processes responsible for ozone formation or less at a particular location. The time of observed maximum value (March–May) of O_3 coincided with the time of maximum temperature ($43\text{--}44^\circ\text{C}$), but the time of the maximum value of O_3 during November–December did not coincided with the maximum temperature ($30\text{--}32^\circ\text{C}$). It is noticed during November–December, the temperatures are comparably low. From these studies, it is pertinent to mention that temperature rise beyond a certain limit is understood to have led to the saturation of photochemical production ozone. Lacies et al. (1990) mentioned that the upper troposphere (8–16 km altitude region) plays the major role in greenhouse effect and maximum is near the tropopause level where a 1 DU increase in ozone produces a temperature increase of about 0.02°K). Hence, surface temperature should warm both in response to a decrease in stratospheric ozone (above 30 km) and an increase in tropospheric ozone (Subbaraya and Lal 1998). At the same time, it can also be expected that there could have been the sufficient concentration of ozone precursors at the time, when temperature could have been comparative low.

Figure 2b shows diurnal variations of mean ozone during the three different seasons, as defined earlier, namely winter (December–February), summer (March–May) and monsoon (June–November). In all, the seasons there are a strong diurnal variation, which becomes more distinct in winter and summer, where as the afternoon values are almost 3 times higher than the nocturnal ones. Daytime ozone mixing ratios are observed to be maximum (about 60 ppbv) during summer and lowest (about 22 ppbv) in the monsoon period. The concentration of ozone during the winter period increased from a minimum value of 25 ppbv at around 0800 hours, to a value of 55 ppbv. However, during the summer season the minimum ozone value was 26 ppbv, which rose to a maximum value of 60 ppbv. Highest ozone concentrations were observed both in winter and summer but not in monsoon months. This indicates that the ozone variation is not only controlled by solar radiation, but also by dynamics. Favorable conditions for photochemical O_3 production are high temperature, high intensity of solar radiation, and sufficiently high concentrations of NO (Naja and Lal 2002). All these conditions are satisfied at our site during the summer months, resulting in the observed broad ozone maximum. There is also the possibility of enhanced transport from the stratosphere during the summer season.

A relatively lower amount of surface ozone was observed during the monsoon months than the winter/summer months. It is worth mentioning that low O_3 levels are noticed in all seasons during morning hours because the lower boundary layer height largely reduces the mixing processes between the ozone-poor surface layer and ozone-rich upper layer. The increase of ozone in the troposphere is the result of increase in human produced ozone precursor emissions. Ozone concentrations are found to be increasing in the free troposphere as well as near the surface in many parts of the northern hemisphere (Naja and Lal 2002; Lal et al., 2000; Varotsos et al. 1994; 2001; Nair et al. 2002; Debaje and Kakade 2006).

5.2 Variation of surface ozone during nighttime and daytime in different months

Figure 3 shows the average annual course of the ozone-mixing ratio for the period 2002–2003 at Anantapur. The data have been grouped according to months to delineate the differences in different months in the nighttime (2000–0800 hours) and daytime (0800–2000 hours) changes of O_3 for the January 2002 to December 2003 period at Anantapur. The monthly average O_3 concentrations show a daytime maximum (55 ± 7 ppbv) in March and minimum (28 ± 3.4 ppbv) in August, and corresponding nighttime maximum (37 ± 7.3 ppbv) and minimum (22 ± 2.3 ppbv) with 1σ standard deviation. Table 1 shows the average surface-ozone concentration rate of change in the daytime (08:00–20:00 hours) and nighttime (20:00–08:00 hours) at Anantapur for the study period. The high O_3 concentrations (48 ppbv) were in March while low concentrations (26 ppbv) occurred in August attributable to variations in precursor gases and influence of changing meteorological parameters. The rate of increase of O_3 is high (1.52 ppbv/h) in March and lower (0.40 ppbv/h) in July. The average rate of increase of O_3 from midnight to midday is 1 ppbv/h. It can be seen from Fig. 3 that nighttime values are low in comparison to that of the daytime ones, whereas the daytime and nighttime cycle exhibit maxima in March–May and October–December, respectively.

It is well known that ozone is an important secondary pollutant in the boundary layer and comes from transport from stratosphere across the troposphere into the boundary layer, or from influx from the free troposphere, or itself. The daytime increase in ozone mixing ratio is basically due to photooxidation of precursor gases, like CO, CH_4 , and other hydrocarbons in the presence of sufficient amount of NO/ NO_x . Observational and modeling studies show that elevated rural ozone levels in summer is the product of long-range transport or ozone precursors and multi-day photochemical production and accumulation of ozone (Guicherit and Van Dop 1977; Varotsos et al. 2001). The temporal and spatial variability of the ozone has been tested over Antarctica region by Varotsos (2005b) using the power law correlations. Currently, the estimation of the scaling exponents of the power law is achieved by the employment of the detrended fluctuation analysis (DFA), which has already proved its usefulness in several complex systems like the surface air-pollutants

Fig. 3 Annual course of the ozone mixing ratios for the period 2002–2003 at the rural station of Anantapur

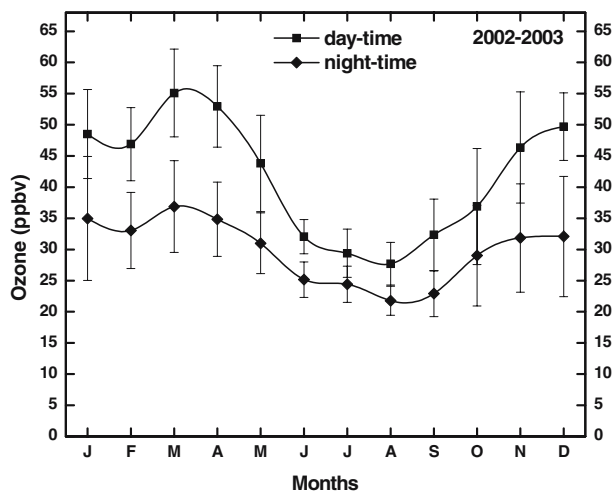


Table 1 The monthly mean O₃ concentrations with 1 σ deviation and O₃ concentrations observed during 20:00–08:00 h (nighttime) and 08:00–20:00 h (daytime) and its increase rate for the period January 2002–December 2003 at rural station of Anantapur

Month	Average O ₃ (20:00–08:00 hours) (Nighttime)	Average O ₃ (08:00–20:00 hours) (daytime)	Increase O ₃ rate (ppbv/h)	Mean \pm 1 sigma
January	34.9 \pm 9.9	48.5 \pm 7.1	1.13	45.6 \pm 7.6
February	33 \pm 6.1	46.9 \pm 5.8	1.15	42.7 \pm 6
March	36.8 \pm 7.3	55.1 \pm 7.0	1.52	48.9 \pm 7.7
April	34.8 \pm 5.9	52.9 \pm 6.5	1.50	46.8 \pm 7.1
May	30.9 \pm 4.8	43.8 \pm 7.7	1.07	39.6 \pm 7.8
June	25.1 \pm 2.8	32 \pm 2.7	0.57	30.4 \pm 2.7
July	24.4 \pm 2.8	29.3 \pm 3.8	0.40	28.1 \pm 3.8
August	21.7 \pm 2.3	27.7 \pm 3.4	0.5	26.2 \pm 3.5
September	22.9 \pm 3.6	32.3 \pm 5.7	0.78	30.4 \pm 6.4
October	29 \pm 8.0	36.8 \pm 9.2	0.65	35.2 \pm 8.9
November	31.8 \pm 8.7	46.3 \pm 8.9	1.20	43.0 \pm 9.3
December	32 \pm 9.6	49.7 \pm 5.4	1.47	46.9 \pm 5.9

(Varotsos et al. 2005), the total ozone content (Varotsos 2005a, b) and the global tropospheric temperature (Varotsos and Kirk-Davidoff 2006). These studies contribute to the development of more reliable simulating models for the ozone, nitrogen oxides and particulate matter fluctuations.

Boundary layer processes and meteorological parameters also play a role in the O₃ variabilities. Mixing ratios of ozone start increasing gradually after sunrise, attaining maximum values during near local noon. After sunrise, boundary layer height gradually increases from about 200–300 m to about 1,500–2,000 m during noon hours due to convective heating, and the layer becomes less stratified. During this period, air at lower levels, which has low amounts of ozone, mixes with the air from greater heights which is relatively rich in ozone. Therefore, average ozone mixing ratios during 12:00–14:00 h could be representative of the upper boundary layer and the free lower tropospheric ozone levels. The photochemical production of ozone is directly connected to the temperature and the solar flux, its concentration increases with respect to both parameters. For instance, transported Peroxyacetyl Nitrate (PAN), anthropogenic carbon monoxide, methane and less reactive hydrocarbons can be considered as responsible for the observed ozone maximum/minimum in the background troposphere and at remote sites away from man-made polluted sources. Low levels of O₃ observed during nighttime is due to the lack of O₃ production from photooxidation of precursors. In addition, the titration of O₃ by NO in the shallow boundary layer and loss due to surface deposition continues during the nighttime. The higher and broad noontime peak during 14:00–16:00 hours appearance is due to higher mixing ratios of NO_x and faster titration.

5.3 Monthly variations in ozone and meteorological parameters

Figure 4a shows the seasonal variation of rural ozone at Anantapur in the form of average monthly values for the 2-year period (2002–2003) is presented. The vertical bars denote 1 σ standard deviation. Minimum values are observed in the months of June, July and August and the maximum values are observed in the months of March and April with a ratio of 1: 2. Similar patterns of seasonal variation have also been observed in the rural sites like

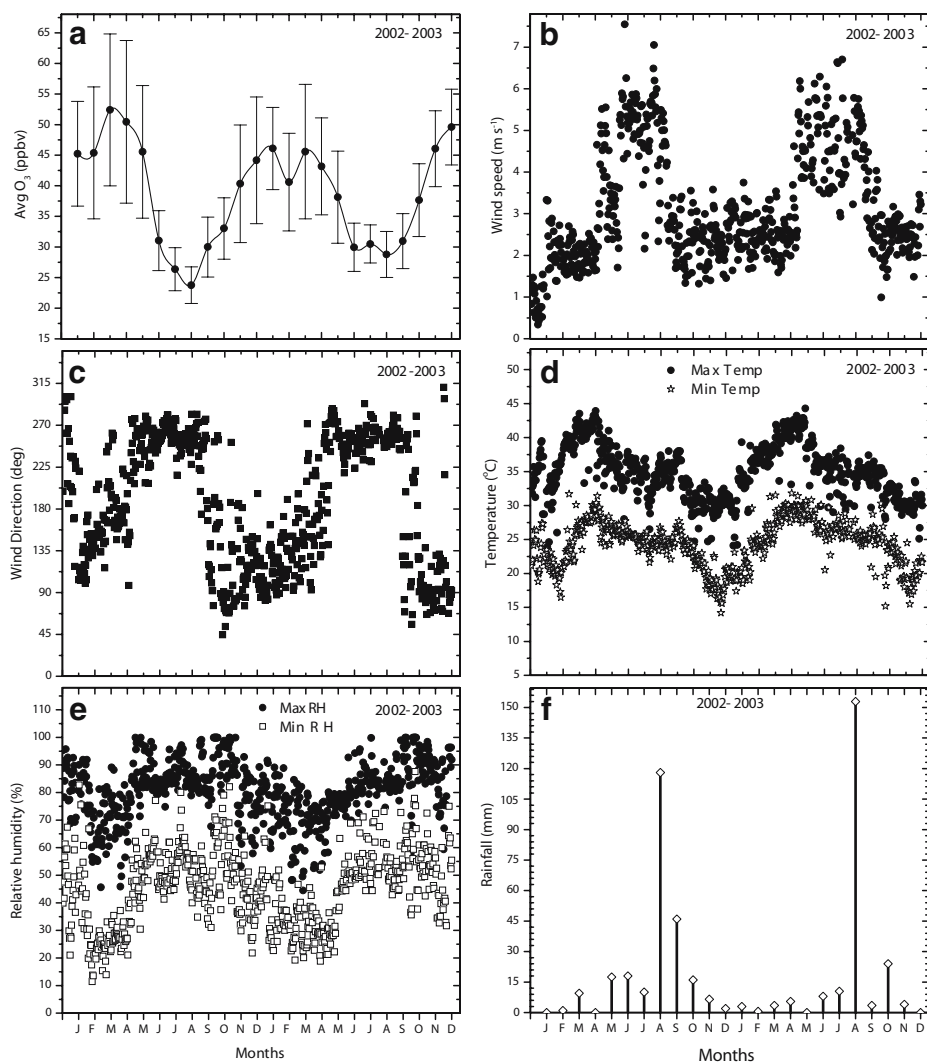
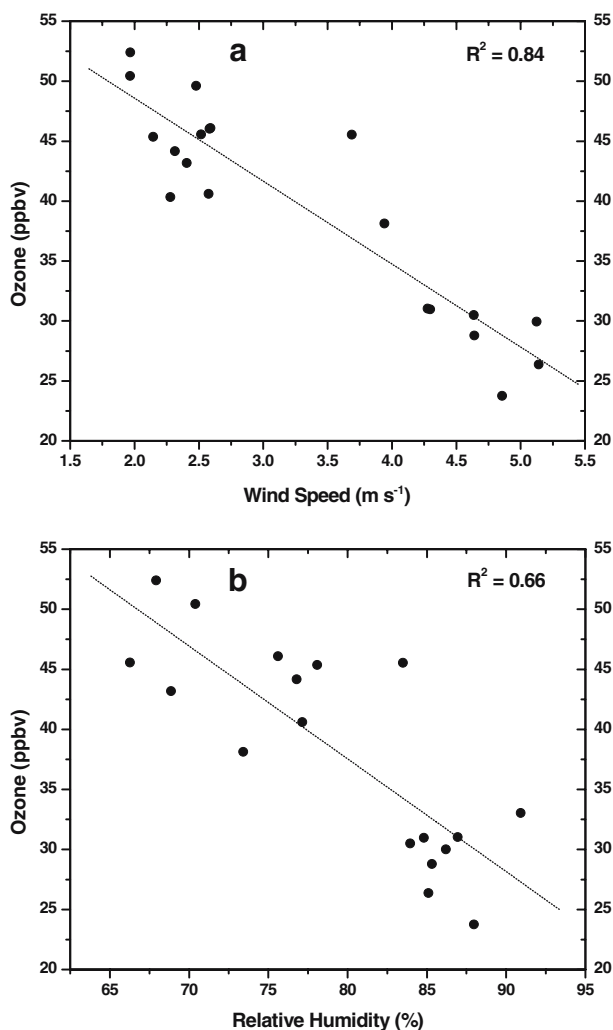


Fig. 4 **a** Seasonal variation of rural ozone at Anantapur during 2002–2003 (monthly mean values), **b** daily average wind speed, **c** daily average wind direction, **d** temperature (maximum and minimum of the day), **e** relative humidity (maximum and minimum of the day) and **f** total rainfall, observed at rural station of Anantapur during 2002–2003

Gadanki in India during the past decade (Naja and Lal 2002). Figure (4b–f) shows monthly variations of wind speed, wind direction, temperature, relative humidity and total rainfall are respectively for the period of observations. Surface temperature is highest (43–44°C) during March and April months leading to higher photochemical production. On the other hand, RH, which is higher during the rainy season, shows negative correlation with temperature and ozone mixing ratio (Nair et al. 2002). There is a satisfactory correlation ($R^2=0.84$) between wind speed and ozone concentration. As seen in earlier cases (Nair et al. 2002), humidity is negatively related with ozone concentration and may be expected to cause reduction in ozone through deposition mechanism. The southwest monsoon, which

starts by the end of May, is quite active during June and July and gets weakened by August. The months of August, September and October recorded with considerable rainfall during this period of measurements. In the monsoon period, the sky is generally overcast, decreasing the solar insolation and thereby reducing the photochemical processes. Also during the rainy season some of the pollutants are washed out. These two factors could cause a low level of ozone during the monsoon period. The meteorological parameters such as temperature, relative humidity, wind speed, wind direction and rainfall do have drastic changes during the year, which resulted in considerable variabilities in O_3 . Figure(5a,b) shows scatter plots of surface ozone, wind speed and relative humidity measured at Anantapur during period of 2002–2003. It can be seen that among the two parameters, measured correlation of O_3 with wind speed is better ($R^2=0.84$) in comparison with the correlation of relative humidity ($R^2=0.66$). The respective correlation coefficients (R^2) are 0.84 and 0.66 respectively.

Fig. 5 Correlations (with coefficient values R^2) between ozone and meteorological parameters(**a** wind speed and **b** relative humidity) observed simultaneously during 2002–2003 at Anantapur, India



6 Summary and conclusions

The annual average diurnal variation of O_3 shows maximum concentration (46 ppbv) at noon and minimum (25 ppbv) in the morning with 1σ standard deviation. The average seasonal variation of ozone mixing ratios are observed to be maximum (about 60 ppbv) during summer and minimum (about 22 ppbv) in the monsoon period. The monthly average high (low) O_3 48.9 ± 7.7 ppbv (26.2 ± 3.5 ppbv) observed at noon in March (August) is due to the possible increase in precursor gas concentration by anthropogenic activity and the influence of meteorological parameters. The rate of increase of surface ozone is high (1.52 ppbv/h) in March and lower (0.40 ppbv/h) in July. The average rate of increase of O_3 from midnight to midday is 1 ppbv/h. Mixing ratios of ozone increase more during the morning than the ozone decrease during the evening. This type of diurnal variation in ozone with mixing ratios remaining high until the late evening has also been observed at some rural sites. This has been attributed to slower titration of ozone by NO during late evening time at rural sites. Mixing ratio of ozone starts increasing at about 0730 h, it reaches to a maximum value at about 16:00 hours. The diurnal peak is rather broad, extending from 11:00 to 18:00 hours. Mixing ratio of ozone starts decreasing after 18:00 hours. The diurnal cycle of O_3 is explained by nighttime O_3 destruction under the formation of a nocturnal inversion layer and by daytime convective boundary layer mixing and photochemical O_3 production. The monthly average O_3 concentrations show a daytime maximum (55 ± 7 ppbv) in March and minimum (28 ± 3.4 ppbv) in August, and corresponding nighttime maximum (37 ± 7.3 ppbv) and minimum (22 ± 2.3 ppbv) with 1σ standard deviation. The daytime increase in ozone mixing ratio is basically due to photooxidation of precursor gases, like CO, CH_4 , and other hydrocarbons in the presence of sufficient amount of NO/NO_x . It is of interest to note that while the diurnal maximum temperature occurred during the afternoon period, when the solar radiation was higher. Highest ozone concentrations were observed both in winter and summer but not in monsoon months. Nighttime values are low compare to that of the daytime ones, whereas the daytime and nighttime cycle exhibit maxima in March–May and October–December, respectively. It is well known that ozone is an important secondary pollutant in the boundary layer and comes from transport from stratosphere across the troposphere into the boundary layer, or from influx from the free troposphere, or itself. Surface temperature is highest (43–44°C) during March and April months leading to higher photochemical production. On the other hand, relative humidity (RH), which is higher during the rainy season, shows negative correlation with temperature and ozone mixing ratio. There is a satisfactory correlation ($R^2=0.84$) between wind speed and ozone concentration.

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